Nonlinear acoustic and microwave absorption in glasses

M. Kirkengen[(1)] and Yu. M. Galperin[(1,2)]

[(1)] *Department of Physics, University of Oslo, P. O. Box 1048 Blindern, N 0316 Oslo, Norway*

[(2)] *A. F. Ioffe Physico-Technical Institute, 194021 St. Petersburg, Russia*

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Abstract

A theory of weakly-nonlinear low-temperature relaxational absorption of acoustic and electromagnetic waves in dielectric and metallic glasses is developed. Basing upon the model of two-level tunneling systems we show that the nonlinear contribution to the absorption can be anomalously large. This is the case at low enough frequencies, $\omega\tau_0(T) \ll 1$, where $\tau_0(T)$ is the minimal relaxation time for two-level systems with the inter-level splitting $\sim k_BT$. In dielectric glasses, the lowest-order nonlinear contribution is proportional to the wave's intensity. It is negative and exhibits anomalous frequency and temperature dependencies, $\Delta\Gamma/\Gamma_0 \propto [\omega\tau_0(T)]^{-1/2}T^{-2}$. In metallic glasses, the nonlinear contribution is also negative, and it is proportional to the *square root* of the wave's intensity and to the frequency. Numerical estimates show that the predicted nonlinear contribution can be measured experimentally.

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I. INTRODUCTION

Acoustic and dielectric properties of glasses were intensively studied during the last decades. The most remarkable result[1] of these studies was the discovery of universal low-temperature properties that are only weakly dependent on the chemical composition of the glass. These "anomalous" specific properties include low-temperature specific heat, thermal conductivity, thermal expansion, propagation of ultrasound, dielectric loss, electric and acoustic echo and some other properties. An important step towards an understanding of the "anomalous" properties of glasses was the introduction of the tunneling model[2,3]. According to this model, there exist *two-level systems* (TLS) associated with local tunneling states in double-well potentials. These states are characterized by an energy difference 2Δ between the minima and a tunnel splitting 2Λ, the energy spacing being

$$2\epsilon \equiv 2\sqrt{\Delta^2 + \Lambda^2}\,. \tag{1}$$

The parameters Δ and Λ are random, the distribution of Δ and $\ln\Lambda$ being assumed constant. There are several review articles[4–8] where experimental data and their interpretation on the basis of the tunneling model are given.

Two different absorption mechanisms are usually discussed in connection with the TLS. The first one is resonant absorption, which is a direct absorption of acoustic (or microwave) quanta $\hbar\omega$ accompanied by transitions between the levels of TLS. Resonant absorption of an acoustic wave can be expressed as[4]

$$\Gamma^{(\mathrm{res})} = \alpha(\omega/s)\,\tanh\left(\hbar\omega/2k_BT\right) \tag{2}$$

where α is a dimensionless coupling constant, s is the sound velocity, while T is the temperature. The case of microwave differs from Eq. (2) only by the coupling constant, so we will discuss below only the case of acoustic waves. At high enough temperature ($\hbar\omega \ll k_BT$) Eq. (2) yields

$$\Gamma^{(\mathrm{res})} \approx \alpha\hbar\omega^2/2sk_BT\,. \tag{3}$$

Another mechanism - *relaxational absorption* - is due to modulation of the populations of the TLS levels by the alternating deformation field created by the acoustic wave. This modulation is due to the time-periodic variation in the inter-level spacing 2ϵ. This variation produces, in its turn, a modulation of the levels' populations which lags in phase the variation of ϵ. This lag leads to the energy dissipation. The *linear* in the acoustic intensity relaxational absorption can be estimated as[4,9]

$$\Gamma^{(\mathrm{rel})} \approx \frac{\alpha}{s} \begin{cases} \tau^{-1}, & \omega\tau_0 \gg 1; \\ \omega, & \omega\tau_0 \ll 1, \end{cases} \tag{4}$$

where $\tau_0(T)$ is the minimum relaxation time of the TLS having $\epsilon \approx k_B T$. Comparing Eq. (2) with Eq. (4) we conclude that the relaxational absorption always predominates at $\omega\tau_0 \ll 1$. If $\omega\tau_0 \gg 1$ the ratio $\Gamma^{(\mathrm{res})}/\Gamma^{(\mathrm{rel})} \approx \hbar\omega^2\tau_0/k_B T$ can be either greater or less than one under experimentally accessible conditions.

Nonlinear absorption was mostly studied for the resonant mechanism. Actually, the nonlinear resonant absorption of sound is one of the basic effects confirming the existence of TLS in disordered materials (see e. g. Ref. 4). The factor $\tanh(\hbar\omega/2k_B T)$ in Eq. (2) has a transparent physical meaning – it is the difference in equilibrium populations of the lower and upper levels of a *resonant* TLS having the inter-level spacing $\hbar\omega$. The reason for the nonlinearity is that intense sound equalizes the levels' populations, which again results in a decrease in the absorption. Experimentally, the effect takes place at very low acoustic intensities[9]. Consequently, the resonant absorption decreases with the intensity increase, and at large enough amplitudes only the relaxational contribution can be observed. As a result, the *nonlinear* relaxational absorption becomes important.

The source of the nonlinear relaxational absorption is a strong modulation of the inter-level spacing of the relevant TLS (with the inter-level distance $\approx k_B T$) by the sound wave. As a result, the relevant TLS are able to absorb energy only during a part of the sound period. Consequently, the total absorption decreases with the sound amplitude. A theory for the nonlinear relaxation absorption in insulation glasses has been developed in[10,11], the authors were concentrated on the case of large acoustic intensities. It seems that it is difficult

to realize such a regime in a realistic experimental situation with insulating glasses. However, the nonlinear relaxational absorption was observed experimentally in metallic glasses (PdSiCu, PdSi, PdNiP)[12] as a specific two-stage nonlinear behavior (successive decrease of resonant and then relaxational absorption as the intensity increases). The relevant theory for the case of large intensities has been developed in Refs. 13.

In this paper we show that one can expect pronounced effects in nonlinear relaxational absorption even at relatively low intensities. Namely, we claim that at low enough acoustic frequencies the nonlinear corrections to the linear absorption coefficient $\Gamma_0^{(\mathrm{rel})}$ are rather pronounced and possess unusual frequency and temperature dependencies. We hope that the regime of "weak nonlinearity" is easier accessible for the experiments, especially with insulating glasses.

According to the theory for the strongly nonlinear regime[10,11], significant deviation from the linear relaxational absorption takes place at $d \gtrsim k_B T$ where d is the amplitude of modulation of the inter-level spacing ($\sim k_B T$) of the relevant TLS. Consequently, one can think that small nonlinear corrections to the linear absorption coefficient behave as $(d/k_B T)^2$. However, the energy difference 2ϵ between the levels depends on both the diagonal splitting Δ and the tunnel coupling Λ according to Eq. (1). Furthermore, the relaxation time τ for a given TLS is also dependent on its parameters Δ and Λ. This dependence can be expressed as (cf with Refs. 4,8)

$$\frac{1}{\tau(\Delta,\Lambda)} = \frac{1}{\tau_0(T)} \left(\frac{\epsilon}{k_B T}\right)^{\beta+1} \left(\frac{\Lambda}{\epsilon}\right)^2 \coth\left(\frac{\epsilon}{k_B T}\right) . \tag{5}$$

The first factor has a meaning of a *minimal* relaxation time for the systems with the inter-level spacing $\epsilon \approx k_B T$. The fact that the inter-level transitions can take place only if the wells are coupled is described by the factor $(\Lambda/\epsilon)^2$. This factor is maximal at $\Delta = 0$ or at $\Lambda = \epsilon$. In the factor $(\epsilon/k_B T)^{\beta+1}$ one power of ϵ is due to deformational coupling between TLS and internal degrees of freedom[4,8], while ϵ^β describes the energy dependence of the density of states for the internal degrees of freedom. In insulating glasses these are phonons ($\beta = 2$) while in metallic glasses the relaxation is due to electronic excitations near the Fermi level

($\beta = 0$)[8]. The last factor represents the sum of the occupation numbers for the excitation responsible for the absorption and emission processes. Indeed, $\coth(\epsilon/k_BT) = 2N(2\epsilon) + 1$ where N is the Planck function. Consequently, nonlinear effects in absorption are determined by the ratio between the amplitude of modulation of the diagonal splitting, d, and the characteristic energy, δ, which is a combination of the parameters Δ and Λ. This combination is determined by the dimensionless product $\omega\tau_0$, and it appears that $\delta \ll k_BT$ at $\omega\tau \ll 1$ (see below). As a result, at low frequencies nonlinear effects must be pronounced. They must also show specific frequency dependence. Having in mind that at $\omega\tau \ll 1$ $\quad \Gamma_0^{(\mathrm{rel})} \propto \omega$ and temperature-independent, one can expect that nonlinear effects can be detected also from anomalous temperature and frequency dependencies of absorption. This is the main message of the paper.

Below we will sketch the basic theoretical approach to calculate both linear and weakly nonlinear absorption. Then we will concentrate on the nonlinear correction to the absorption coefficient for relatively low amplitudes, both for insulating and metallic glasses.

II. THEORETICAL BASIS

The Hamiltonian of a TLS in the external ac field can be written as[9]

$$\mathcal{H} = (\Delta + d\,\cos\omega t)\sigma_z - \Lambda\sigma_x\,, \tag{6}$$

where $\sigma_{x(z)}$ are Pauli matrices, Δ is the energy gap between the levels of the isolated potential wells, while Λ is the tunnel matrix element, $d = \gamma_{ik}u_{ik}^{(0)}$ in the case of sound wave (γ_{ik} is the deformational potential of the TLS, $u_{ik}^{(0)}$ is the amplitude value of the deformation tensor). In the case of electromagnetic wave $d = \eta\mathcal{E}_0$, where η is the dipole moment of the TLS while $\mathcal{E}_0$ is the amplitude of the electric field. The corresponding change in the tunneling transmittance Λ we shall assume to be small and neglect[9,8].

According to the TLS model[2,3] we assume Δ and Λ to be independent random quantities, Δ and $\ln\Lambda$ being uniformly distributed over a broad range of values in comparison with the

temperature. We shall also assume the deformational potential γ_{ik} to be a random quantity, uncorrelated with Δ and Λ , the distribution of which has a maximum (a similar assumption can be introduced concerning η).

Following Refs. 10,11, we consider the case of relatively low frequencies when the energy of the acoustic quantum $\hbar\omega$ is much less that the characteristic inter-level distance δ of the TLS which make a contribution to the nonlinear absorption (we shall estimate δ later). At $\hbar\omega \ll \delta$ one can employ the adiabatic approximation and neglect time derivatives of the external field while solving the Schrödinger equation for the TLS. In this approximation the TLS is characterized by the time-dependent spacing $2\epsilon(t)$,

$$\epsilon(t) = \sqrt{(\Delta + d\cos\omega t)^2 + \Lambda^2}\,, \tag{7}$$

and the occupation numbers of the upper (n) and the lower $(1-n)$ levels. The non-equilibrium occupation numbers can be found from the balance equation

$$\frac{dn}{dt} = -\frac{n - n_0(t)}{\tau(t)} \tag{8}$$

where $\tau(t)$ and $n_0(t)$ are given by the substitution $\epsilon(t) \to \epsilon$ into the Eq. (5) and into the expression

$$n_0 = [\exp(2\epsilon/k_BT) + 1]^{-1} \tag{9}$$

for the equilibrium occupation number. According Eq. (8), the dynamics of the levels' population is characterized by a time-dependent relaxation time. The solution of Eq. (8) for a strongly nonlinear regime ($d \gg k_BT$) has been analyzed in Refs. 10,11 for the case of insulating glasses and in Ref. 13 for metallic glasses. Below we shall present a theory for the regime of weak nonlinearity which seems easier to realize.

III. CALCULATIONS

In the following we will use the subscript 0 to indicate linear results. The power absorbed by a single TLS can be determined by the expression[10]

$$p(\Delta,\Lambda)=\frac{\omega}{\pi}\int_0^{2\pi/\omega} n\frac{d\epsilon}{dt}\,dt \tag{10}$$

The contributions of individual TLS must be added and such a summation can be performed in a conventional way using the distribution function of the random parameters Δ and Λ and replacing the deformational potential γ_{ik} by its average value. The distribution function is usually[2,3] chosen as

$$N(\Delta,\Lambda)=2N/\Lambda$$

where N is the the density of states per energy interval while $1/\Lambda=d(\ln\Lambda)/d\Lambda$ describes a smooth distribution of $\ln\Lambda$. The coefficient 2 is introduced because we use the notation 2ϵ for the energy interval. This gives for the total absorption

$$P=2N\int_0^\infty d\Delta\int_0^\infty\frac{d\Lambda}{\Lambda}\,p(\Delta,\Lambda,d)\,. \tag{11}$$

To analyze the absorption one can use the exact periodic in time solution of Eq. (8) to obtain[10]

$$\begin{aligned}P=\frac{N}{\Theta}\int_0^\infty\int_0^\infty\frac{d\Delta\,d\Lambda}{k_BT\,\Lambda}\left(1-e^{-\int_0^\Theta dt_1/\tau(t_1)}\right)^{-1}\\ \times\int_0^\Theta\int_0^\Theta\frac{dt\,dt'\,\dot\epsilon(t)\dot\epsilon(t-t')}{\cosh^2[\epsilon(t-t')/k_BT]}e^{-\int_0^{t'}dt_1/\tau(t-t_1)}.\end{aligned} \tag{12}$$

Here $\Theta=2\pi/\omega$ is the period of the wave. We will first consider the case of dielectric glasses. In the linear approximation one has

$$\dot\epsilon_0(t)=-(\Delta/\epsilon)\,\omega d\sin\omega t\,,\quad \epsilon=\sqrt{\Delta^2+\Lambda^2}\,,$$

while the relaxation rate is time-independent and given by Eq. (5). From Eq. (12) one obtains the well-known expression for the relaxational absorption[9]

$$P=\frac{N\omega}{2k_BT}\int_0^\infty\int_0^\infty\frac{d\Delta\,d\Lambda}{\Lambda\,\cosh^2(\epsilon/k_BT)}\,\frac{\omega\tau}{1+\omega^2\tau^2}\,. \tag{13}$$

Since $\tau(\Delta,\Lambda)\propto\Lambda^{-2}$ the typical value of Λ in this integral is k_BT at $\omega\tau_0\gg1$ and $k_BT\sqrt{\omega\tau_0}$ at $\omega\tau_0\ll1$. It is the last regime that produces anomalous nonlinear effects. In the linear case at $\omega\tau_0\ll1$ one obtains

$$P_0 = (\pi^2/16) N \omega d^2 \tag{14}$$

As we shall see, the nonlinear corrections to the absorption are of greatest importance for low values of the inter-level spacing $\epsilon \ll k_B T$. Consequently, one can still use the asymptotic expression for $\coth(\epsilon/k_B T) \approx k_B T/\epsilon$ in Eq. (5). As a result, in the case of $\beta = 2$ (i. e. for insulating glasses) $\tau = \tau_0 \, (k_B T/\Lambda)^2$, and it is *time-independent*. That leads to important simplifications in Eq. (12). Indeed, to get nonlinear corrections to the absorption one has to expand only the function

$$\dot{\epsilon}(t) = -\frac{\Delta + d\cos\omega t}{\sqrt{(\Delta + d\cos\omega t)^2 + \Lambda^2}} \, \omega d \, \sin\omega t$$

in powers of d up to second order, and then perform the integrations over t, t', Δ and Λ. Such a calculation can be easily performed using any computer algebra package (we used "Maple"). The result can be expressed as

$$(P - P_0)/P_0 = -c_d \, (d/k_B T)^2 \, [\omega \tau_0(T)]^{-1/2} \, . \tag{15}$$

Here $c_d = (3/32)(4\sqrt{2} - 1) \approx 0.44$ is the numerical factor.

In the case of metallic glasses, the linear calculations are performed analogous to the dielectric case. Making use of the Λ dependence of the relaxation time that emphasizes small Λ we get $P_0 \approx 0.38 N \omega d^2$ for the most interesting case $\omega\tau \ll 1$. For the nonlinear corrections, however, we can no longer assume the transition time to be time-independent. Indeed, calculations show that the main contribution to the nonlinear absorption comes from the expansion of the exponential in Eq. (12). Expanding the power p absorbed by a single TLS up to the fourth order in the wave's amplitude and subtracting the quadratic contribution p_0 (which is responsible for the linear absorption coefficient) we obtain

$$p - p_0 = \frac{\nu \, (\nu \sin\theta - 8) \, \pi \sin^2\theta \cos^4\theta}{(4 + \nu\sin\theta)\,(1 + \nu\sin\theta)} \, \frac{d^4}{\epsilon^2} \, .$$

Here we denote $\nu = (\omega\tau_0)^{-1}$, $\Lambda = \epsilon \sin\theta$, $\Delta = \epsilon\cos\theta$.

As can be seen, this expression diverges strongly for small ϵ, so it is not possible to obtain an exact quantitative estimate from our simplified calculation. In order to make an order-of

magnitude-estimate, however, we may cut off the integral in the lower limit, at $\epsilon = |d|$. This assumption yields for $\omega\tau \ll 1$

$$(P - P_0)/P_0 = -c_m(\omega\tau_0)^{-1}\,(|d|/k_BT) \tag{16}$$

with c_m is of the order 1. We observe that the nonlinear contribution is proportional to $|d|$, i. e. to the *square root* of the intensity rather than to the intensity, as in usual cases. This dependence arises from the fact that nonlinear effects are especially important for the TLS with small inter-level splitting $\epsilon \lesssim |d|$. Indeed, these systems are strongly perturbed by the acoustic wave. However, the probability to find a TLS with small splitting is proportional to $|d|$. Furthermore, at small ϵ the relaxation time the $\tau_0(\epsilon)$ increases as ϵ^{-1} that leads to a decrease of the contribution of the low-energy TLS. Collecting of all the factors leads to the estimate (16). Unfortunately, our calculation based on the expansion in powers of d/ϵ cannot provide the numerical factor. However, it yields the proper functional dependencies.

In the high-frequency limit, $\omega\tau \gg 1$, the nonlinear contribution is $\propto (d/k_BT)^2$.

IV. DISCUSSION

Since $P_0 \propto \omega$ and independent of T, the nonlinear contribution to the sound absorption in the *dielectric glasses* appears $\propto \sqrt{\omega}$, the temperature dependence (at fixed d) being $\propto T^{-2}[\tau_0(T)]^{-1/2} \propto T^{-1/2}$.

Analyzing the relevant integrals, one comes to the conclusion that the characteristic inter-level spacing of the TLS responsible for the absorption is $\delta \sim k_BT(\omega\tau_0)^{1/4} \leq k_BT$. This fact confirms that one can employ low-energy asymptotics in Eq. (5) and replace $\cosh^{-2}(\epsilon/k_BT) \to 1$ in Eq. (12). Inequality $\hbar\omega \ll \delta$ appears met for realistic parameters of the glass.

Now, let us give a rough estimate of the intensities needed to obtain a significant nonlinear contribution for dielectric glasses. One can estimate τ_0 as $(\hbar/k_BT)(T_c/T)^2$ where the constant T_c depends on the interaction constant between TLS and thermal phonons.

As for the most glasses $T_c \sim 20$ K we get at $T = 0.1$ K $\tau_0 \approx 0.3 \cdot 10^{-7}$ s. Thus at $\omega/2\pi = 50$ kHz $\omega\tau_0 \approx 10^{-2}$. We assume the deformational potential $\gamma_{ik} \approx 1$ eV and express the amplitude of the deformation tensor $u_{ik}^{(0)}$. The relation between the acoustic intensity W and the deformation tensor can be expressed as $u_{ik}^{(0)} \approx W/(\rho s^3)$ (where s is the sound velocity, while ρ is the density of the glass).

Demanding that the nonlinear correction should be about 10% of the absorption we get

$$\begin{aligned} W \ \left(\mathrm{W/cm^2}\right) &= 0.1\,(\omega\tau_0)^{1/2}\,(\rho s^3/c_d)\ (k_B T/\gamma)^2 \\ &\approx 0.03\,[T\,(\mathrm{K})/0.1]^{1/2} \end{aligned} \tag{17}$$

for $\rho = 5$ g/cm^3, $s = 3 \cdot 10^5$ cm/s.

In the case of metallic glasses we get $\tau_0 \approx \hbar/\chi T$ with $\chi \approx 0.01$. This gives a total temperature dependence of T^{-2} compared to $T^{-1/2}$ in the dielectric case. The characteristic interlevel spacing in the case of metallic glasses is $\delta = (d\,k_B T\,\omega\tau_0)^{1/2} \ll k_B T$.

Using the same values for γ, ρ and s for the case of metallic glasses we get

$$W\ (\mathrm{W/cm^2}) \approx \rho s^3 \omega\tau_0\,(0.1 k_B T/\gamma)^2 \approx 0.01\,[T\,(\mathrm{K})/0.1]^2\,.$$

To conclude, we have estimated nonlinear contributions to low-temperature absorption of acoustic waves and microwave in dielectric and metallic glasses. It is shown that at low enough frequencies, $\omega\tau_0(T) \ll 1$, they are anomalously large and can be detected experimentally. In the high frequency regime, $\omega\tau_0(T) \geq 1$ the nonlinear contributions are proportional to the intensity and do not exhibit anomalous frequency and temperature dependencies.

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